

III. Summary and Conclusions

Through a combination of ground based measurements in different locations during different seasons new details regarding single particle mixing state were discovered. A key element leading to these findings was the development of the mobile laboratory that was used 6 of the 8 principle studies conducted during the course of this grant. The mobile laboratory allowed for rapid and flexible deployment while lowering the barriers to sampling in locations with varying availability of power.

The list of objectives accomplished during this study included: investigations into time and size resolved chemical composition detailing short term changes in composition, monitoring spatial variability of particulate matter and sources, seasonal and interannual variability in particulate matter size and composition, mass comparisons and investigations into the volatile and semivolatile fraction of particles, analyzing changes in particle composition linked with meteorology and air mass back trajectories, and development of source apportionment algorithms. With respect to short term changes in composition two chapters (Chapters 1 and 2) demonstrate the ability of the ATOFMS to measure ultrafine particles and ship plumes on the time scale of a few minutes. This increase in time resolution allowed us to detect and characterize processes that would not have been possible previously. In the future higher time resolution will allow for greater investigations into the transformative processes that convert fresh aerosols to aged background particles.

Investigations into spatial variability were made possible by the ability to move site to site multiple times within a day, as made possible by the mobile laboratory. Chapter 3 describes mobile laboratory results showing the difference between a clean background day and a moderately polluted day. In the two cases for San Diego both spatial and temporal (diurnal) patterns were observed, with temporal patterns playing a larger role in the accumulation mode with spatial and temporal being roughly equal within the ultrafine mode. Future investigations focusing on ultrafine particles will link well with the high time resolution capabilities described above to monitor source to background transitions.

One of the primary objectives of this report was to investigate seasonal and interannual differences in aerosol size and chemical composition within California. Chapter 4 focused on SOAR I and II in 2005 provides a detailed analysis of the change from diurnal patterns in the summer to buildup periods in the fall over several days. A more detailed analysis comparing seasonal differences in the chemistry of alkylamines is described in Chapter 5. By analyzing particle volatility ATOFMS was able to show that acidic particle cores in summer led to the formation of aminium nitrate and sulfate salts, while less acidic cores during the fall led to more volatile amine species. Linking the chemistry to volatility demonstrates the utility of exploring further systems using the thermodenuder-ATOFMS system discussed. Chapter 6 details the changes in particle size and chemistry interannually and finds that the summers of 2005, 2006, and 2007 had considerable differences between them. The chapter then discusses how best to characterize the chemistry observed and simplify it for incorporation into future modeling efforts.

A key advance with respect to quantifying ATOFMS measurements has been the ability to convert to mass concentrations from the raw counts measured by the ATOFMS.

A method to quantitatively scale the data using an aerodynamic particle sizer (APS) is described in Chapter 7 and compared with standard mass measurements including the micro-orifice uniform deposit impactor (MOUDI) and beta attenuation monitor (BAM). Variation within the calculations used for this conversion are discussed in detail in Chapter 8 and when taken together demonstrate the ability to present ATOFMS data on both a number and mass basis. This method is used in both Chapter 3 and Chapter 10 to present meaningful comparisons to standard mass measurements in the field and in trailer and will be used frequently in future analysis.

The intercomparison of ATOFMS data with meteorology and air mass back trajectories for the purpose of understanding sources and secondary contributors to particulate matter (PM) concentrations was a key focus of this report. The impact of trace metals in the particle phase on human health has received considerable attention in recent years and in Chapter 9 meteorology and air mass history are used to predict source regions for a plethora of metals in the urban atmosphere of Riverside, CA. Transported aerosols (including those containing metals such as vanadium) have also been studied in detail. The impact of transported emissions from the Los Angeles Port region on San Diego are discussed in detail in Chapter 10, where it is shown that port sources can make a significant contribution to local PM in San Diego. The impact of transported emissions during unique episodes such as the 2007 San Diego wildfires were also studied as a part of this report and Chapter 11 describes both the types of particles observed as well as their ability to act as cloud condensation nuclei (CCN), which has important implications for clouds and precipitation.

Significant advances have been made in our ability to apportion the sources of different particles in the atmosphere. Chapter 12 ties our initial dynamometer studies that were conducted as part of a previous CARB grant with field measurements to determine the fraction of cars and trucks in an urban environment. The methods that originated with car/truck differentiation are then expanded to incorporate major sources of PM_{2.5} in Chapter 13. These two chapters provide detailed information both on the location studied, but also led to development that will allow for future analysis of sources from a more mathematical perspective. Lastly, the issue of aging and its impact on source apportionment are discussed in Chapter 14. The ability to determine the sources of aged particles in critical in a polluted urban environment and considerable advances in our ability to apportion this quantitatively are shown.

Together the research described in this report depicts a number of advances in field capabilities, analysis capabilities, as well as the answers to many of the complex scientific questions posed in the initial proposal. In the future single particle analysis has considerable potential to provide information on mixing state with impacts ranging from source apportionment to climate and precipitation patterns.

IV. References

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V. Appendices

Publications and Presentations

1. Publications

- 1) Qin, X. Y.; Bhave, P. V.; Prather, K. A., Comparison of two methods for obtaining quantitative mass concentrations from aerosol time-of-flight mass spectrometry measurements. *Anal. Chem.* 2006, 78, (17) 6169-6178.
- 2) Shields, L. G.; Qin, X.; Toner, S. M.; Prather, K. A., Detection of ambient ultrafine aerosols by single particle techniques during the SOAR 2005 campaign. *Aerosol Sci. Technol.* 2008.
- 3) Toner, S. M.; Shields, L. G.; Sodeman, D. A.; Prather, K. A., Using mass spectral source signatures to apportion exhaust particles from gasoline and diesel powered vehicles in a freeway study using UF-ATOFMS. *Atmos. Environ.* 2008, 42, (3) 568-581.
- 4) Pratt, K. A.; Prather, K. A., Real-time, single-particle volatility, size, and chemical composition measurements of aged urban aerosols. *Environ. Sci. Technol.* 2009, In Preparation.
- 5) Ault, A. P.; Moore, M. J.; Furutani, H.; Prather, K. A., Impact of emissions from the Los Angeles port region on San Diego air quality. *Environ. Sci. Technol.* 2009, 43, (10) 3500-3506.
- 6) Pratt, K. A.; Hatch, L. E.; Prather, K. A., Seasonal volatility dependence of ambient particle phase amines. *Environ. Sci. Technol.* 2009, 43, (14) 5276-5281.
- 7) Ault, A.; Gaston, C.; Wang, Y.; Dominguez, G.; Thiemens, M. H.; Prather, K., Characterization of the Single Particle Mixing State of Individual Ship Plume Events measured at the Port of Los Angeles. *Environ. Sci. Technol.* 2009, submitted.
- 8) Toner, S. M.; Moffet, R. C.; Dall'Osto, M.; Harrison, R. M.; Prather, K. A., Source apportionment of PM_{2.5} in Athens (Greece) and Mexico City using an ATOFMS derived mass spectral source library. *Atmos. Environ.* in prep.
- 9) Toner, S. M.; Shields, L. G.; Prather, K. A., Source Apportionment of freeway-side PM_{2.5} using ATOFMS. *Atmos. Environ.* in prep.
- 10) Shields, L. G.; Qin, X.; Toner, S. M.; Prather, K. A., Aging effects on source apportionment from the single particle perspective. *Atmos. Environ.* in prep.
- 11) Shields, L. G.; Qin, X.; Toner, S. M.; Prather, K. A., Characterization of tracemetalts in single urban particles during the SOAR 2005 campaign. *Science of the Total Environment* in prep.
- 12) Qin, X.; Shields, L. G.; Toner, S. M.; Prather, K. A., Single Particle Characterization in Riverside, CA during the SOAR 2005 Campaign – Part 1: Seasonal Comparisons. *Environ. Sci. Technol.* in prep.
- 13) Qin, X.; Prather, K. A., The Effect of APS Scaling functions on the Quantification of Aerosol Time-of-Flight Mass Spectrometry. *Atmos. Environ.* in prep.
- 14) Yandell, M. A.; Ault, A. P.; Pratt, K. A.; Wang, Y.; Zauscher, M.; Gaston, C. J.; Prather, K. A., Air pollution transport from the Ports of Long Beach and Los

- Angeles and its impacts on the local community. *Aerosol Sci. Technol.* 2009, In Prep.
- 15) Creamean, J. M.; Pratt, K. A.; Ault, A. P.; Qin, X.; Gaston, C. J.; Shields, L. G.; Prather, K. A., Inter-annual Comparison of Real-Time, Single-Particle Ambient Aerosol Measurements Using ATOFMS in Riverside, CA. 2009, in prep.
 - 16) Ault, A. P.; Creamean, J. M.; Prather, K. A., Mobile Laboratory Observations of Temporal and Spatial Variability within the Coastal Urban Aerosol. *Aerosol Sci. Technol.* in prep.

2. Presentations

- 1) **Qin, X.Y.**; Bhave, P.V.; Prather, K.A.; ATOFMS single particle measurements and quantification. ACS Spring 2005 National Meeting San Diego, CA March, **2005**
- 2) **Shields L.G.**, Suess D.T., Prather K.A. Determination of single particle mass spectral signatures from heavy duty vehicle emissions in the 0.1 to 3 micrometer size range. ACS Spring 2005 National Meeting San Diego, CA March, **2005**
- 3) **Toner, S.M.**, Shields, L.G., Sodeman, D.A., Prather, K.A.; Using mass spectral source signatures to apportion exhaust particles from gasoline and diesel powered vehicles in a freeway study. ACS Spring 2005 National Meeting San Diego, CA March, **2005**.
- 4) **Prather, K.A.**; Sullivan, R.C; Guazzotti, S.; Denkenberger, K.A.; Holecek, J.; Moffet, R.C.; Spencer, M.T.; Qin, X.Y.; Using the temporal variability of single particle mixing state to understand aging, heterogeneous chemistry, and the sources of atmospheric particles. ACS Spring **2006** National Meeting
- 5) **X. Qin** and K. A. Prather. "Direct Measurements of Ambient Aerosols Mixing State and Source Apportionment Using Single Particle Mass Spectrometry", Invited Talk, United States Department of Agriculture, Manhattan, KS, March, 2007.
- 6) **X. Qin** and K. A. Prather. "Direct Measurements of Ambient Aerosols Mixing State Using Single Particle Mass Spectrometry", Invited Talk, NanoScale Corporation, Manhattan, KS, March, 2007.
- 7) **Ault, Andrew P.**; Dominguez, G.; Furutani, H.; Thiemens, M.; Prather, K A Mass Spectral Fingerprint of Ship Emission Particles by Aerosol Time-of-Flight Mass Spectrometry. *AAAR 26th Annual conference*, Reno, NV, United States, September 24-28, **2007**.
- 8) **Ault, Andrew**; Gaston, Cassandra, Wang, Ying, Kimberly Prather Observation and Characterization of Individual Ship Plumes by Aerosol Time-of-Flight Mass Spectrometry. *25th Informal Symposium on Kinetics and Photochemical Processes in the Atmosphere*, UCLA, Los Angeles California, February 19, **2008**.
- 9) **Ault, A.**; Gaston, C; Wang, Y.; Zauscher, M.; Moison, R; Prather, K; Single Particle Observations of Variability within the Urban Aerosol. *AAAR 27th Annual Conference*, Orlando, FL, United States, October 20-24, **2008**.
- 10) **Yandell, Margaret A.**; Ault, Andrew P.; Pratt, Kerri A.; Wang, Ying; Zauscher, Melanie; Gaston, Cassandra J.; Prather, Kimberly A. Air pollution transport from the Ports of Long Beach and Los Angeles and its impacts on the local community. *26th Informal Symposium on Kinetics and Photochemical Processes in the*

Atmosphere. Informal Conference on Atmospheric Kinetics University of California, Riverside, **2009**.

- 11) **Ault, A.P.**; Creamean, J.M.; Prather, K.A.; Measurements of Spatial Variability in the Urban Atmosphere With Aerosol Time-of-Flight Mass Spectrometry. *AAAR 28th Annual Conference*, Minneapolis, MN, United States, October 26-30, **2009**